

Interactive comment on “Large Contributions from Biogenic Monoterpenes and Sesquiterpenes to Organic Aerosol in the Southeastern United States” by Lu Xu et al.

Anonymous Referee #2

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The manuscript presented by Xu et al. proposes an interesting study on the contribution of the oxidation of alpha-pinene and caryophyllene to the SOA mass observed in the S.E.-US. The characterization of SOA generated in the lab-in-the-field smog chamber was performed using an aerosol mass spectrometer. Overall the work performed in this study is good and fall within the scope of the journal. However, I think the conclusions proposed from the PMF analysis/chamber experiments are not always well sustained and more caution should be taken when extrapolating the results.

General comments: The authors should carefully review their paper and avoid the repetition between the main text and the SI. At many places, sentences are duplicated

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and are not useful. However, some important details are left within the SI and should be moved to the main manuscript.

The authors should provide more information in the PMF analysis and provide the elementary checks to validate their analysis. For instance, it is a bit surprising that the factors don't change throughout the experiments (i.e. bf vs af) while significant perturbation has been made to the system. Or do the authors consider/claim that most of the SOA sampled in the ambient are formed from the oxidation of alpha-pinene or caryophyllene? In addition, we could expect that the fresh LO-OOA (formed within a few minutes, without lights) would have different signatures than LO-OAA formed in the atmosphere (aged SOA, formed from different chemistry, ...). How do the factors correlate throughout the experiments: e.g. LO-OOA_Amb_Bf vs LO-OOA_Chamber_Af? How do the identified factors correlate with the reference MS? How do the residuals evolve throughout an experiment? How does alpha-pinene-derived LO-OOA correlate with caryophyllene derived LO-OOA? Overall, the authors should provide more statistical analyses in order to give a robust validation of the analysis.

The authors should report the concentration of the inorganics in their experiments and in case of significant concentrations of sulfate estimate the aerosol acidity. Indeed, the presence of acidic aerosols can lead to multiphase reactions (e.g. reactive uptake of IEPOX) that could greatly impact the SOA composition. In addition, an estimation (modeling?) of the concentrations of other VOCs would be interesting (especially isoprene). Ozonolysis of alpha-pinene leads to the formation of OH radicals, which could further react and oxidize alpha-pinene but also other VOCs present in the ambient air. The authors should discuss this possibility and provide more information in the background of the chamber/ambient air. As it is, the conclusions proposed in the paper on the potential increase of the IEPOX-OA or COA factors from the oxidation of alpha-pinene and caryophyllene, respectively appear speculative (correlations are not sufficient to validate such trend: $r \sim 0.5$). For instance, the authors could estimate the amount of IEPOX (thus isoprene) formed in the chamber to explain the formation of

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IEPOX-OA and check if the numbers make sense or not.

Specific comments: Lines 104:111. Did the authors characterize the chamber? Mixing, wall losses,...

Lines 141: The authors claim that by having an overflow, it suppressed the particle loss. Did they mean reduce? Have you done some tests to validate such statement?

Lines 217-220: Why not using the outdoor chamber to do such experiments? Can the authors discuss the strategy here?

Lines 266-268: The decay of LO-OOA is quite fast and I do not think it can only explain by the dilution and or dead-volume. The residence time in the chamber is ~ 100 min. Where were located the sampling inlets?

Lines 278:284: It is quite expected. What is the point of the authors?

SI Line 150:157: These results are a bit intriguing. The data reported for the boreal forest do not exhibit prominent ions at m/z 53 or 82. The authors suggest that alpha-pinene/monoterpene can contribute to IEPOX-OA but according to Fig S7 the correlation is far to be obvious strong. The authors should compare the MS obtained in their study with other PMF data obtained from monoterpene-dominated areas (e.g. boreal forest).

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